

## PATENT

### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

| Applicant: Luis Mendez Llatas, et al.             | RE: PRELIMINARY AMENDMENT      |
|---|--------------------------------|
| Serial No.: Unknown                               | Date: August 8, 2001           |
| Filed: Simultaneously herewith                    | Examiner: Unknown              |
| For: "HETEROGENEOUS CATALYST ) COMPONENTS FOR " ) | Group Art Unit: Unknown        |
| )   | Our Ref: 618993-3/JP/B-3406DIV |

Hon. Commissioner of Patents and Trademarks Box Patent Application Washington, D.C., 20231

Sir:

Before issuing the first Office Action on the merits, please enter the following amendments and remarks into the prosecution history of the above-identified U.S. patent application without prejudice.

# IN THE SPECIFICATION

Please replace the first paragraph on page 13 of the specification (see lines 1-7 on page 13) with the amended paragraph set forth below. (Appendix A, which is enclosed herewith, shows how the first paragraph on page 13 was amended to produce the amended paragraph set forth below. In Appendix A, the portions being added are underlined; and the portions being deleted are enclosed in brackets.)

P: NH<sub>2</sub>, NHR, SH, OH or PHR

V+z+w=3, v being different from 0

t and u are comprised between 0 and 10.

Some examples of compounds of formula V are:

3-Mercaptopropyltrimethoxysilane, 3aminopropyltrimethoxysilane, N-Phenylpropyltrimethoxysilane,
N-Methylpropyltrimethoxysilane, NAminopropyldimethoxymethylsilane, 3-mercaptopropyltrimethoxysilane.

# IN THE CLAIMS

Please cancel Claims 1-13 without prejudice.

Please add the following new claims.

--Claim 14. A heterogeneous metallocene catalyst component obtained by reacting a porous inorganic support containing hydroxyl groups with a metallocene compound, wherein the metallocene compound has a formula (I):

$$\left[\begin{array}{c} Y_{j} - (E)_{0} - Z \\ k \end{array}\right]_{k}^{L} MY_{(11+2)}$$

(I)

wherein:

Y is halogen;

 ${f M}$  is a transition metal of group 3, 4, 5, or 6 of the Periodic Table of the Elements;

each  ${\bf L}$  is independently a cyclopentadienyl-type unity, wherein the cyclopentadienyl-type unity is optionally substituted with substituents, wherein the substituents are equal or different, and wherein each  ${\bf L}$  is united to  ${\bf M}$  through a  $\pi$  bond;

Z is a group that forms a union bridge between the two unities L, wherein Z has 0 to 20 carbon atoms and 0 to 5 oxygen, sulfur, nitrogen, phosphorus, silicon, germanium, tin or boron atoms; E is a group having 0 to 20 carbon atoms and 0 to 5 oxygen, sulfur, nitrogen, phosphorus, silicon, germanium, tin or boron atoms, wherein E has a skeleton, and wherein the skeleton has at least one silicon, germanium or tin atom, and wherein Y is united with the silicon, germanium, or tin atom of E;

o is a number of value 0 or 1;

k is a number of value 1, 2 or 3;

**m** is a number equal to or higher than 2 and coinciding with an oxidation state of the transition metal;

 ${f j}$  is a number of value 0 or 1; wherein when  ${f j}$  is 1 and  ${f o}$  is 0,  ${f Z}$  has at least one silicon, germanium or tin atom which  ${f Y}$  is directly united to; wherein each  $Y_j-(E)_o-Z$  group is the same or different, and wherein  ${f j}$  is equal to 1 in at least one  $Y_j-(E)_o-Z$  group; provided that the metallocene compound does not have general formula

$$\mathbf{X_{m}},\ \mathbf{M^{1}}\left(\mathbf{L'-M^{2}}\left(\mathbf{R^{1}R^{2}}\right)-\mathbf{A'-Z'R^{3}_{o}},\mathbf{Hal_{p}},\right)_{\mathbf{n}},,$$

wherein  $\mathbf{M}^1$  is a metal of group 4, 5 or 6 of the periodic table, each  $\mathbf{X}$  is independently hydrogen, halogen or a  $C_1$ - $C_{40}$  carbon-containing rest;  $\mathbf{m}'$  is equal to 1, 2 or 3;  $\mathbf{n}'$  is equal to 1 or 2; each  $\mathbf{L}'$  is independently a pi ligand, wherein each  $\mathbf{L}'$  coordinates to central atom  $\mathbf{M}^1$ ; each  $\mathbf{M}^2$  is independently silicon, germanium or tin;  $\mathbf{R}^1$  is a  $C_1$ - $C_{20}$  carbon-containing group;  $\mathbf{R}^2$  is a  $C_1$ - $C_{20}$  carbon-containing group or a pi ligand, wherein each  $\mathbf{R}^2$  coordinates to central atom  $\mathbf{M}^1$ ; each  $\mathbf{A}'$  is independently a divalent  $C_1$ - $C_{40}$  carbon-containing rest; each  $\mathbf{Z}'$  is independently boron, silicon, germanium or tin; each  $\mathbf{R}^3$  is independently hydrogen or a  $C_1$ - $C_{20}$  carbon-containing rest;  $\mathbf{o}'$  is equal to 0, 1 or 2; each  $\mathbf{Hal}$  is independently a halogen atom; and  $\mathbf{p}'$  is equal to 1, 2 or 3.

Claim 15. A catalyst component as claimed in claim 14, wherein the metallocene compound has a formula (II):

$$\begin{bmatrix} Y_{j} & (A)_{n} \\ (R)_{p} & (A)_{l} \end{bmatrix}_{k}^{L} MY_{(m-2)}$$

(II)

wherein:

each Y is halogen;

 ${\bf M}$  is a transition metal of group 3, 4, 5 or 6 of the periodic table;

each  ${\bf L}$  is independently a cyclopentadienyl-type unity, wherein the cyclopentadienyl-type unity is optionally substituted with substituents, wherein the substituents are equal or different, and wherein each  ${\bf L}$  is united to  ${\bf M}$  through a  $\pi$  bond;

 ${f Q}$  is an element of group 13, 14 or 15;

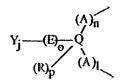
 ${f E}$  is a group having 0 to 20 carbon atoms and 0 to 5 oxygen, sulfur, nitrogen, phosphorus, silicon, germanium, tin or boron atoms, wherein  ${f E}$  has a skeleton, and wherein the skeleton has at least one silicon, germanium or tin atom, and wherein  ${f Y}$  is united with the silicon, germanium, or tin atom of  ${f E}$ ;

**R** is hydrogen, halogen, halocarbon, substituted halocarbon,  $C_1$ - $C_{20}$  alkyl,  $C_2$ - $C_{20}$  alkenyl,  $C_6$ - $C_{20}$  aryl,  $C_7$ - $C_{40}$  alkylaryl,  $C_7$ - $C_{40}$  arylalkyl,  $C_8$ - $C_{20}$  arylalkenyl, alkoxy, siloxy, or a combination thereof;

each **A** is equal to or different from each other, wherein each **A** is a bridge group between unities **L** and **Q**, wherein each **A** is a divalent atom of group 16, a trivalent monosubstituted element of group 15, a tetravalent disubstituted element of group 14, or a chain of 2 or more atoms that are substituted or not;

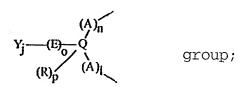
- o is a number of value 0 or 1;
- k is a number of value 1, 2 or 3;
- **m** is a number equal to or higher than 2 and coinciding with an oxidation state of the transition metal;
- p is a number of value 0 or 1;
- n is a number of value 0 or 1;
- 1 is a number of value 0 or 1;
- ${\bf j}$  is a number of value 0 of 1; wherein when  ${\bf j}$  is 1 and  ${\bf o}$  is 0,  ${\bf Q}$  is a silicon, germanium or tin atom;

wherein each



group is the

same or different and wherein j is equal to 1 in at least one



provided that the metallocene compound does not have general formula

$$X_m$$
,  $M^1(L'-M^2(R^1R^2)-A'-Z'R^3_o, Hal_p,)_n$ ,

wherein  $\mathbf{M}^1$  is a metal of group 4, 5 or 6 of the periodic table, each  $\mathbf{X}$  is independently hydrogen, halogen or a  $C_1$ - $C_{40}$  carbon-containing rest;  $\mathbf{m}'$  is equal to 1, 2 or 3;  $\mathbf{n}'$  is equal to 1 or 2; each  $\mathbf{L}'$  is independently a pi ligand, wherein each  $\mathbf{L}'$  coordinates to central atom  $\mathbf{M}^1$ ; each  $\mathbf{M}^2$  is independently silicon, germanium or tin;  $\mathbf{R}^1$  is a  $C_1$ - $C_{20}$  carbon-containing group;  $\mathbf{R}^2$  is a  $C_1$ - $C_{20}$  carbon-containing group or a pi ligand, wherein each  $\mathbf{R}^2$  coordinates to central atom  $\mathbf{M}^1$ ; each  $\mathbf{A}'$  is independently a divalent  $C_1$ - $C_{40}$  carbon-containing rest; each  $\mathbf{Z}'$  is independently boron, silicon, germanium or tin; each  $\mathbf{R}^3$  is independently hydrogen or a  $C_1$ - $C_{20}$  carbon-containing rest;  $\mathbf{o}'$  is equal to 0, 1 or 2; each  $\mathbf{Hal}$  is independently a halogen atom; and  $\mathbf{p}'$  is equal to 1, 2 or 3.

Claim 16. A catalyst component as claimed in claim 14, wherein the metallocene compound has a formula III:

$$Y-T$$
 $(A)_{n}$ 
 $MY_{(m-2)}$ 

III

wherein:

**R** is hydrogen, halogen, halocarbon, substituted halocarbon,  $C_1$ - $C_{20}$  alkyl,  $C_2$ - $C_{20}$  alkenyl,  $C_6$ - $C_{20}$  aryl,  $C_7$ - $C_{40}$  alkylaryl,  $C_7$ - $C_{40}$  arylalkyl,  $C_8$ - $C_{20}$  arylalkenyl, alkoxy, siloxy, or a combination thereof;

n is a number of value 0 or 1;

1 is a number of value 0 or 1;

each A is equal to or different from each other, wherein each A is a bridge group between unities L and T, wherein each A is a divalent atom of group 16, a trivalent monosubstituted element of group 15, a tetravalent disubstituted element of group 14, or a chain of 2 or more atoms that are substituted or not; and T is silicon, germanium or tin.

Claim 17. A catalyst component as claimed in claim 14, wherein the metallocene compound has a formula IV:

$$Y-E$$
 $(A)_{n}-L$ 
 $MY_{(n-2)}$ 

IV

wherein:

 $\bf R$  is hydrogen, halogen, halocarbon, substituted halocarbon,  $\rm C_1-C_{20}$  alkyl,  $\rm C_2-C_{20}$  alkenyl,  $\rm C_6-C_{20}$  aryl,  $\rm C_7-C_{40}$  alkylaryl,  $\rm C_7-C_{40}$ 

arylalkyl,  $C_8-C_{20}$  arylalkenyl, alkoxy, siloxy, or a combination thereof;

**E** is a group having 0 to 20 carbon atoms and 0 to 5 oxygen, sulfur, nitrogen, phosphorus, silicon, germanium, tin or boron atoms, wherein **E** has a skeleton, and wherein the skeleton has at least one silicon, germanium or tin atom, and wherein **Y** is united with the silicon, germanium, or tin atom of E;

n is a number of value 0 or 1;

1 is a number of value 0 or 1;

each  ${\bf A}$  is equal to or different from each other, wherein each  ${\bf A}$  is a bridge group between unities  ${\bf L}$  and  ${\bf C}$ , wherein each  ${\bf A}$  is a divalent atom of group 16, a trivalent monosubstituted element of group 15, a tetravalent disubstituted element of group 14, or a chain of 2 or more atoms that are substituted or not; and  ${\bf Q}$  is silicon, germanium, or tin.

Claim 18. A catalyst component as claimed in claim 14, wherein the inorganic support has been previously modified through reaction with a compound having a formula V:

$$\begin{array}{c|c}
\begin{pmatrix} R \\ z \end{pmatrix}_{z} & R \\
\downarrow Si - \left(O - Si\right)_{t} - \left(R\right)_{u} - P \\
\begin{pmatrix} R \\ R \end{pmatrix}_{w} & R
\end{array}$$

#### wherein:

each R is hydrogen, halogen, halocarbon, substituted halocarbon,  $C_{1-20}$  alkyl,  $C_{2-20}$  alkenyl,  $C_{6-20}$  aryl,  $C_{7-40}$  alkylaryl,  $C_{7-40}$  arylalkyl,  $C_{8-20}$  arylalkenyl, alkoxy, siloxy, or a combination thereof;

X is halogen or group OR;
P is NH<sub>2</sub>, NHR, SH, OH or PHR;
v+z+w=3, v being different from 0;
t is 0 to 10:
and u is 0 to 10.

Claim 19. A catalyst component as claimed in claim 18, wherein the inorganic support is selected from the group consisting of silica, silicates, carbonates, phosphates, clays, metal oxides, and mixtures thereof.

Claim 20. A process for preparing a catalyst component as claimed in Claim 14, wherein the metallocene compound and the inorganic support are put in contact by using tetrahydrofurane as a solvent.

Claim 21. A process for polymerization of alpha-olefins, optionally in combination with a cyclic olefin and/or a diene,

wherein the polymerization is catalyzed by the catalyst component claimed in claim 14.

- Claim 22. A process as claimed in claim 21, wherein the process polymerizes monomers selected from the group consisting of ethylene, propene, 1-butene, 1-hexene, 4-methyl-1- pentene, 1-octene, and mixtures thereof.
- Claim 23. A process as claimed in claim 21, wherein the process copolymerizes ethylene in combination with a comonomer selected from the group consisting of propene, 1-butene, 1-hexene, 4-methyl-1-pentene, 1-octene, cyclic olefins, and mixtures thereof.
- Claim 24. A catalyst system as claimed in claim 14, wherein the cyclopentadienyl-type unity is cyclopentadienyl, indenyl, or fluorenyl.
- Claim 25. A catalyst system as claimed in claim 15, wherein the cyclopentadienyl-type unity is cyclopentadienyl, indenyl, or fluorenyl.
- Claim 26. A catalyst system as claimed in claim 16, wherein the cyclopentadienyl-type unity is cyclopentadienyl, indenyl, or fluorenyl.
- Claim 27. A catalyst system as claimed in claim 15, wherein, for **A** the divalent atom of group 16 is -O-; the trivalent monosubstituted element of group 15 is >NR; the tetravalent

disubstituted element of group 14 is  $>C(R)_2$  or  $>Si(R)_2$ ; or the chain of 2 or more atoms that are substituted or not is -C-C-, -C-Si-, -Si-Si-, -Si-O-, -C-O-, -C-N-, -C-C-C-, -C-Si-C-, or -Si-O-Si-.

Claim 28. A catalyst component as claimed in Claim 14, wherein the metallocene compound is selected from the group consisting of ((chloromethylsilanediyl)bis(cyclopentadienyl))zirconium(IV) dichloride and ((chloromethylsilanediyl)bis(cyclopentadienyl))-hafnium(IV)dichloride.

Claim 29. A catalyst component as claimed in Claim 14, wherein the metallocene compound is selected from the group consisting of

and 
$$Y = \begin{cases} R & Cp \\ R & Si \end{cases}$$
  $Y = \begin{cases} R & Cp \\ R & Cp \end{cases}$   $Y = \begin{cases} R & Cp \\ R & Cp \end{cases}$ 

wherein:

 ${\bf M}$  is a transition metal of group 3, 4, 5, or 6 of the periodic table;

 $\bf R$  is hydrogen, halogen, halocarbon, substituted halocarbon,  $\rm C_1-\rm C_{20}$  alkyl,  $\rm C_2-\rm C_{20}$  alkenyl,  $\rm C_6-\rm C_{20}$  aryl,  $\rm C_7-\rm C_{40}$  alkylaryl,  $\rm C_7-\rm C_{40}$  arylalkyl,  $\rm C_8-\rm C_{20}$  arylalkenyl, alkoxy, siloxy, or a combination thereof;

each  $\mathbf{C_p}$  is independently selected from the group consisting of a cyclopentadienyl ring, a substituted cyclopentadienyl ring, a substituted indenyl ring, a nonsubstituted indenyl ring, a substituted fluorenyl ring, and a nonsubstituted fluorenyl ring;  $\mathbf{Y}$  is halogen; and

C is carbon. --

#### REMARKS

Please consider this Preliminary Amendment on the merits before issuing the first Office Action on the merits.

This Preliminary Amendment cancels Claims 1-13 without prejudice and adds new Claims 14-29. Upon amendment, the above-identified patent application will have one independent claim (new Claim 14) and 16 total claims (new Claims 14-29). Therefore, no fees for excess claims are due.

Support for new Claim 14 can be found in, inter alia, the originally filed versions of Claims 1 and 5. Support for new Claims 15 and 17 can be found in, inter alia, the originally filed versions of Claims 2 and 4, respectively. Support for new Claims 14, 15, and 17 is also described below in the next three paragraphs.

First, new Claim 14 is drafted to omit the limitation that E is a spacer group that unites Z and Y because E does not function as a spacer group if j is zero. Similarly, new Claim 15 is drafted to omit the limitation that E is a spacer group that unites Q and Y because E does not function as a spacer group if j is zero. In addition, new Claim 17 is drafted to omit the limitation that E is a spacer group.

Next, new Claim 14 is drafted to specify that each  $Y_j$ -(E) $_o$ -Z group is the same or different and to specify that j is equal to 1

in at least one  $Y_j$ - $(E)_o$ -Z group. Similarly, new Claim 15 is drafted to

specify that each

$$Y_{j}$$
  $(E)_{0}$   $(A)_{n}$   $(A)_{n}$   $(A)_{n}$ 

group is the

same or different and to specify that j is equal to 1 in at least

one

$$Y_j$$
— $(E)_{o}$   $Q$   $(A)_{l}$ 

group.

Support for drafting new Claims 14 and 15 as described in the preceding paragraph can be found in, inter alia, the originally filed versions of Claims 1 and 2, the three formulas in the second line of formulas on page 8 of the specification, and the first and third formulas in the third line of formulas on page 8 of the specification.

Then, new Claim 17 is drafted to change  ${\bf C}$  to  ${\bf Q}$  in formula IV because formula II in the originally filed version of Claim 2 includes a  ${\bf Q}$  rather than a  ${\bf C}$ . Similarly, new Claim 17 is drafted

to specify that  $\mathbf{Q}$ , rather than  $\mathbf{C}$ , is silicon, germanium, or tin. Support for drafting new Claim 17 as described in the preceding sentence can be found in, inter alia, the originally filed version of Claim 2, which specifies that  $\mathbf{Q}$  is silicon, germanium, or tin.

Support for new Claims 16 and 18-23 can be found in inter alia, the originally filed versions of Claims 3, 6, 7, and 10-13, respectively.

Next, support for new Claims 24-26, which recite limitations found in the originally filed version of Claim 1, can be found in, inter alia, the originally filed version of Claim 1. Support for new Claim 27, which recites limitations found in the originally filed version of Claim 2, can be found in, inter alia, the originally filed version of Claim 2.

Support for new Claim 28 can be found in, inter alia, lines 5-6 on page 13 of the specification, lines 8-9 on page 14 of the specification, and lines 23-24 on page 15 of the specification. Support for new Claim 29 can be found in, inter alia, line 25 on page 6 of the specification through line 3 on page 9 of the specification.

Finally, this Preliminary Amendment amends the first paragraph on page 13 of the specification to correct an inadvertent typographical error. As someone with ordinary skill in the art would know, the formulas set forth in lines 5-7 on page 13 of the specification are examples of compounds of formula V, not examples of compounds of formula III.

It is submitted that the above-identified U.S. application is in condition for allowance. Allowance of the application at an early date is solicited.

This Preliminary Amendment cancels Claims 1-13 without prejudice and adds new Claims 14-29. The cancellations and additions described in the preceding sentence were done to claim the scope of the invention that the Applicants elect to claim and were not done to overcome the prior art, obviousness-type double-patenting rejections, rejections under 35 U.S.C. § 112, or any other rejections or objections. The cancellations and additions described in the first sentence of this paragraph shall not be considered necessary to overcome the prior art, obviousness-type double-patenting rejections, rejections under 35 U.S.C. § 112, or any other rejection or objection.

The Applicant reserves the right to seek protection for any unclaimed subject matter either subsequently in the prosecution of the present case or in a divisional or continuation application.

The Commissioner is authorized to charge any additional fees which may be required or credit overpayment to Deposit Account No. 12-0415. In particular, if this Preliminary Amendment is not timely filed, then the Commissioner is authorized to treat this Preliminary Amendment as including a petition to extend the time period pursuant to 37 C.F.R 1.136(a) requesting an extension of time of the number of months necessary to make this response

timely filed; and the petition fee due in connection therewith may be charged to Deposit Account No. 12-0415.

Respectfully submitted,

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90036

#### APPENDIX A

Re: Luis Mendez Llatas, et al.

"Heterogeneous Catalyst Components for . . ."

Our Ref.: 618993-3/JP/B-3406DIV

Date: August 8, 2001

Please replace the first paragraph on page 13 of the specification (see lines 1-7 on page 13) with the amended paragraph set forth below.

P: NH2, NHR, SH, OH or PHR

v+z+w=3, v being different from 0

t and u are comprised between 0 and 10.

Some examples of compounds of formula [III]  $\underline{V}$  are:

3-Mercaptopropyltrimethoxysilane, 3-

aminopropyltrimethoxysilane, N-Phenylpropyltrimethoxysilane,

N-Methylpropyltrimethoxysilane, N-

Aminopropyldimethoxymethylsilane, 3-mercaptopropyltrimethoxy-

silane.